# Supporting Information

Unprecedented CO$_2$ uptake over highly porous N-doped activated carbon monoliths prepared by physical activation

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## Preparation of the precursor mesoporous PAN monolith

Polyacrylonitrile (PAN, procured from Sigma-Aldrich) is taken as the precursor for the preparation of porous polymer monolith with a bicontinuous structure. The monoliths are developed by thermally induced phase separation (TIPS) technique, which involves solubilization of PAN by heating followed by phase separation on cooling. PAN is insoluble in water but soluble in mixture of water and DMSO. In a typical run, 80 mg of PAN is dispersed in 1 mL of 15:85 volume ratio of water and DMSO and heated at 353 K. After the solid is dissolved under magnetic stirring for 4 h, the homogeneous precursor gel obtained is cooled at 293 K. The clear solution gradually turns cloudy and phase separation of the polymer takes place. The
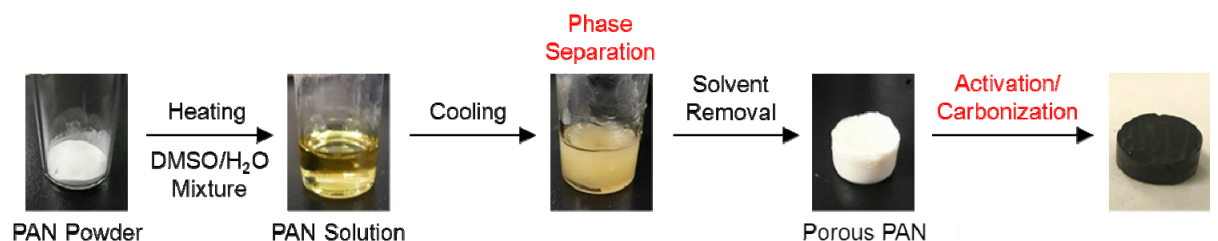

Preparation of the mesoporous PAN monolith by the TIPS

olution separates into two phases with one phase mainly consistingof the mixture of water and DMSO while the other phase is a monolith of PAN. The monolith is recovered, washed repeatedly with methanol to remove the embedded solvent molecules and subsequently dried under vacuum at room temperature. The formation of monoliths is highly dependent on the concentration of the polymer and volume percentage of DMSO. This polymer monolith is subsequently used as a precursor to produce N-doped activated carbon in the form of monolith by a two-step thermal treatment (Scheme 1). The monolith is at first cured by heating in a flow of air at 503 K for 1 h before carbonization. In the next step it is carbonized in a tubular furnace under flow of gas, either Ar or a mixture of 3:1 Ar and CO$_2$ *via* heating ramp of 5° per min from room temperature to the desired temperature (Table 1). The temperature is hold at the highest temperature for 1-2 h, as the case may be, for the different activated carbon samples, before they are cooled down to room temperature.
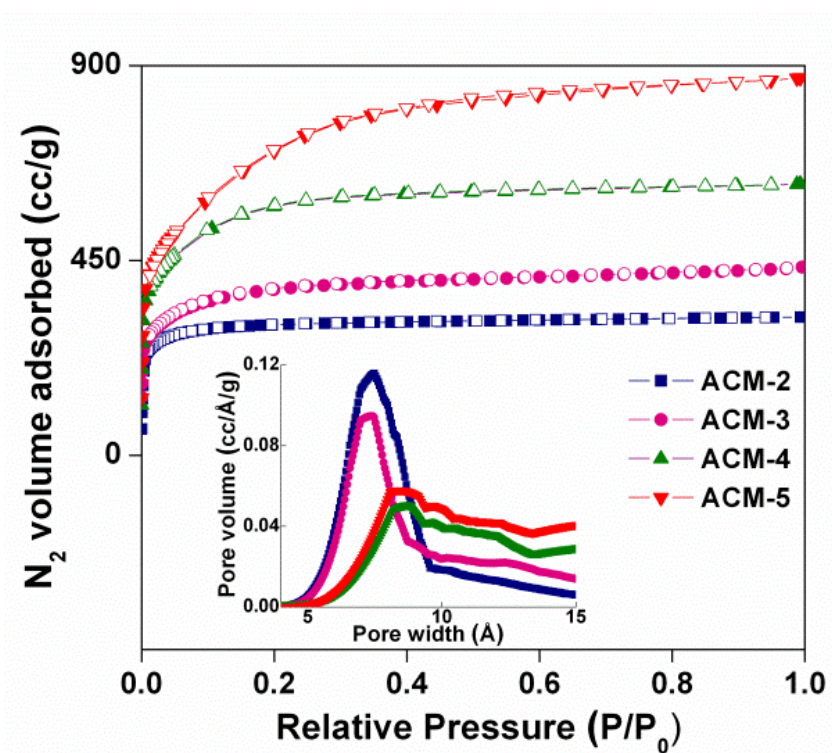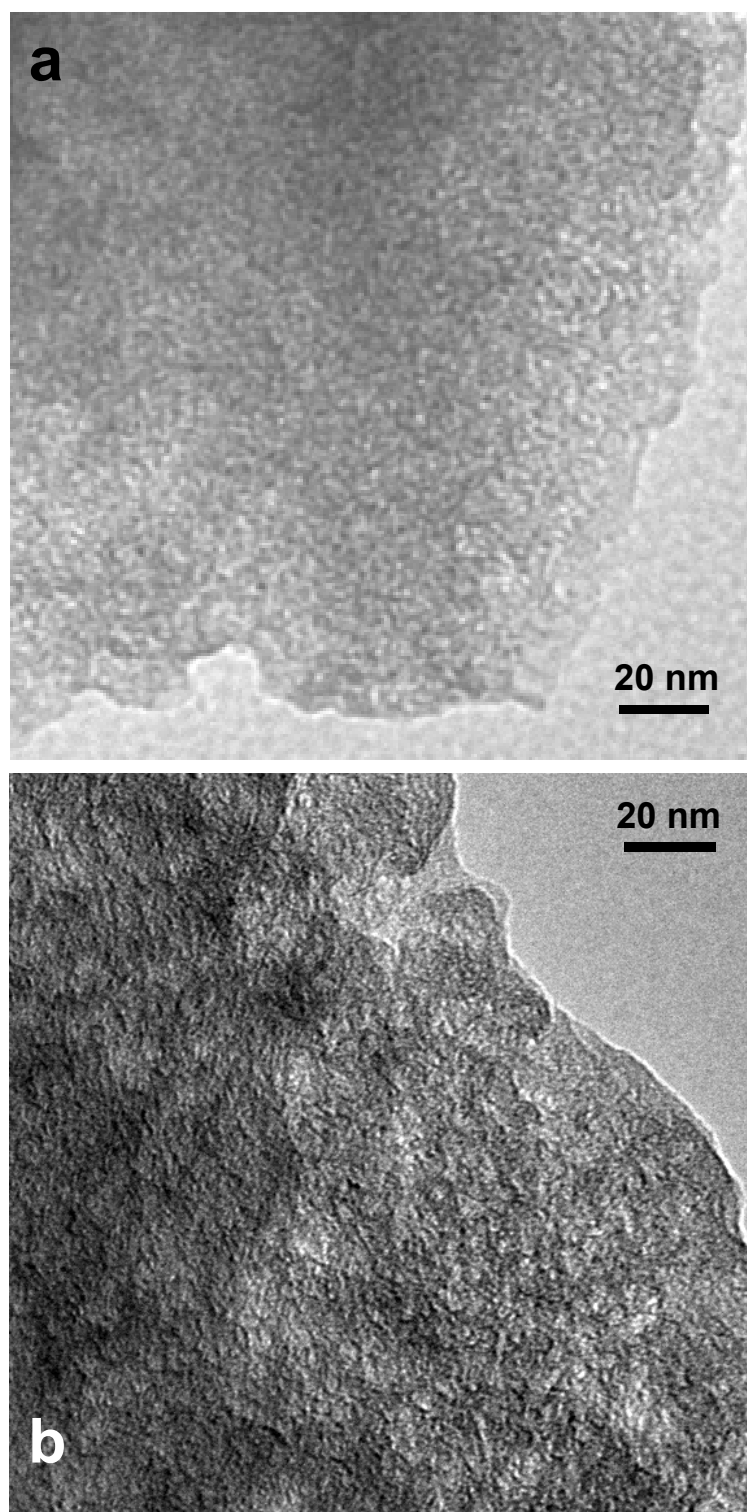
Figure S1: N_2 sorption isotherms of different activated carbon monoliths: adsorption (filled symbol) and desorption (empty symbol). Inset: Pore size distribution using HK method.
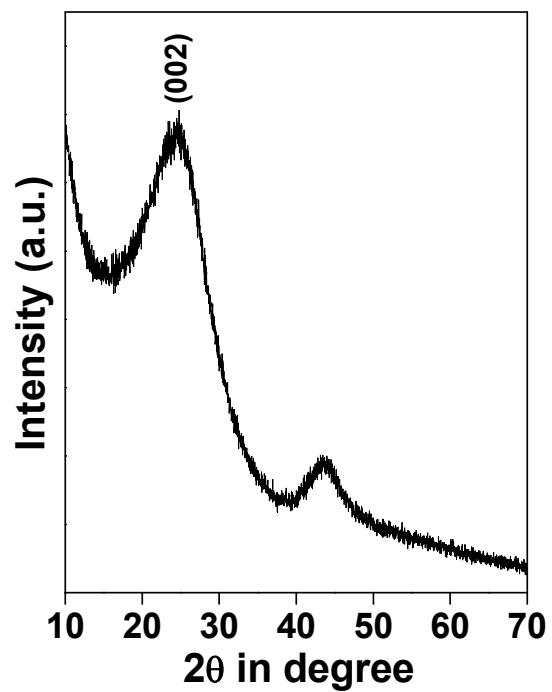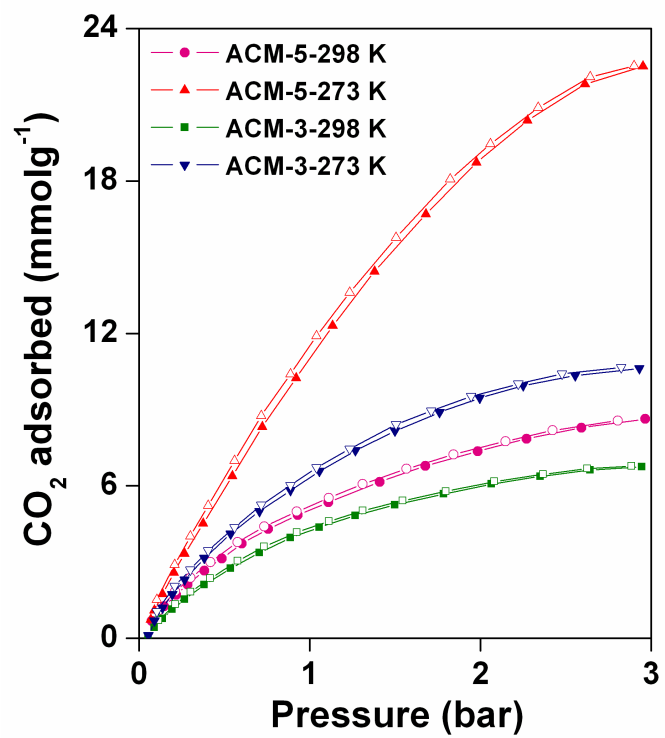
Figure S2: High angle powder X-ray diffraction pattern of **ACM-3**, as a representative case.
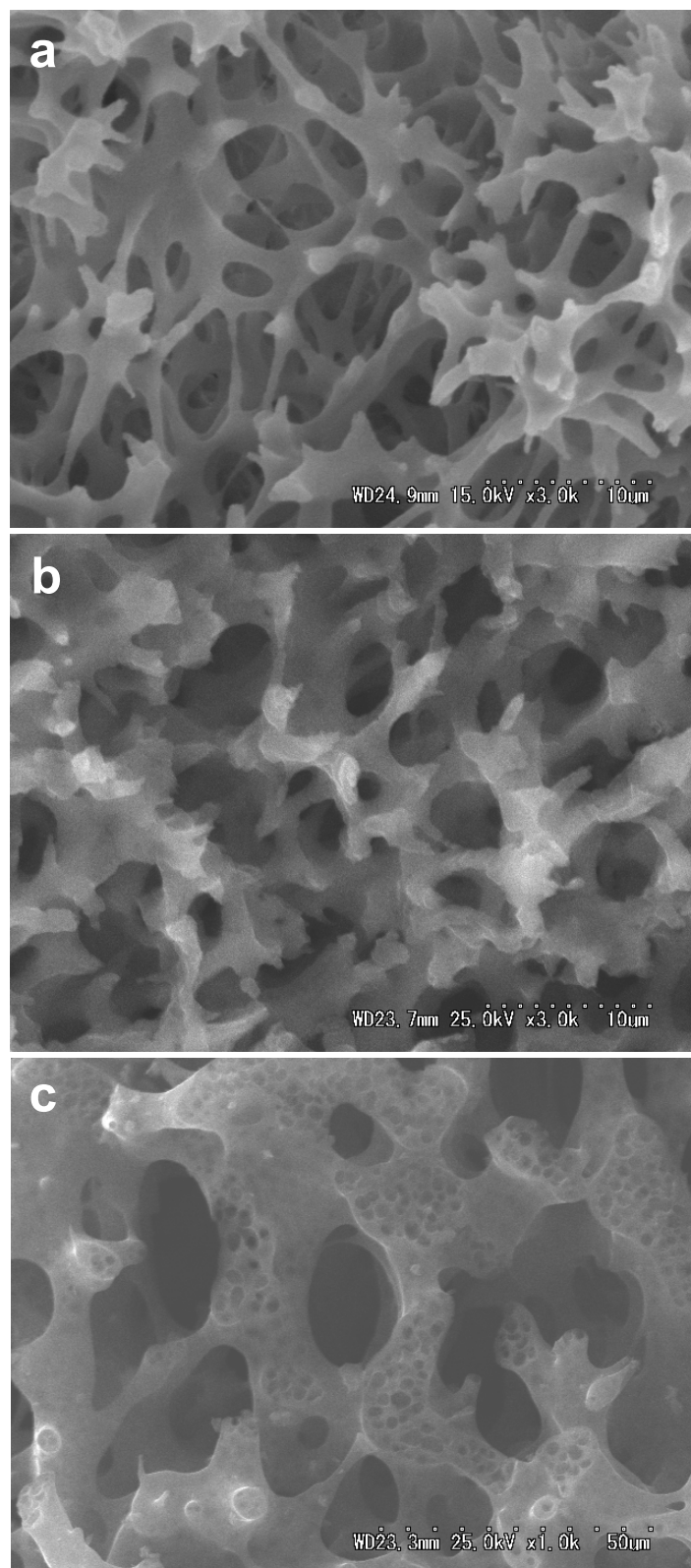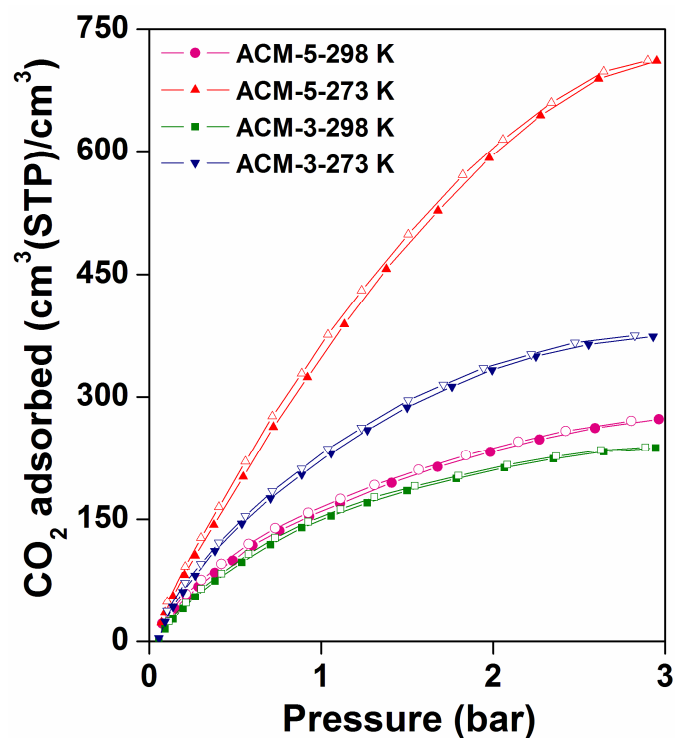
Figure S3: Scanning electron microscopic (SEM) images of the PAN monolith (a), **ACM-3** (b) and **ACM-5** (c).

Figure S4: Transmission electron microscopic (TEM) images of **ACM-3** (a) and **ACM-5** (b).

Figure S5: High pressure CO_2 sorption isotherms of **ACM-3** and **ACM-5** at 273 K and 298 K.

Figure S6: High pressure CO_2 sorption isotherms in a volumetric basis[1] (cm^3/cm^3) of **ACM-3** and **ACM-5** at 273 K and 298 K. (Considering bulk densities of **ACM-3** and **ACM-5** to be 1.57 and 1.41 g/cm^3 respectively).

Reference

1. A. R. Millward and O. M. Yaghi, *J. Am. Chem. Soc.*, 2005, **127**, 17998–17999.